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Relevant to Performance as a Wasteform
in a Potential Tuff Repository

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LWR SPENT FUEL CHARACTERISTICS
RELEVANT TO PERFORMANCE AS A WASTEFORM
IN A POTENTIAL TUFF REPOSITORY

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The Nevada Nuclear Waste Storage Investigations Project (NNWSI) is studying the suitability of the welded devitrified Topopah Spring tuff at Yucca Mountain, Nye County, Nevada, for potential use as a high level nuclear waste repository. The candidate repository horizon is above the water table. However, some water will likely infiltrate the rock. NNWSI spent fuel containers are being designed to meet the 10 CFR 60 requirement⁽¹⁾ that containment of all radionuclides within the waste package be "substantially complete for a period ... not less than 300 years nor more than 1000 years after closure" of the geological repository (the containment period). The 10 CFR 60 requirement for the post-containment period is that "the release rate of any radionuclide ... shall not exceed one part in 100,000 per year of the inventory of that radionuclide calculated to be present at 1000 years following permanent closure" of the repository.

During the post-containment period a limited amount of water could enter a breached container and contact the fuel. The exact condition of the fuel rods at this time is not easily predicted. A majority of the rods are expected to be intact, but a significant number of rods may be breached, containing fine holes or cracks in the cladding. As time progresses, additional fuel rod degradation may occur as a result of cladding corrosion, or splitting of the cladding as fuel within breached rods slowly oxidizes and swells. A testing program has been initiated to determine the probable condition of spent fuel during the post-containment period under NNWSI site specific conditions, and to determine relevant radionuclide release rates for spent fuel. The current testing program is focused on three subject areas: 1) spent fuel leaching/dissolution behavior, 2) spent fuel oxidation and 3) cladding corrosion.

The initial spent fuel leaching/dissolution tests used unoxidized segments of PWR fuel rods and were conducted in air at ambient hot cell temperature. (Use of partially oxidized fuel and elevated temperature are planned for future testing). Four test specimen configurations representing a range of potential degrees of cladding degradation were tested in both deionized water and in J-13 reference NNWSI groundwater; these were:

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- 1) bare fuel removed from split cladding plus the split cladding,
- 2) fuel rod segments containing a 150 micron wide by 2.5 cm long slit through the cladding,
- 3) fuel rod segments containing two approximately 200 micron diameter laser drilled holes through the cladding, and
- 4) fuel rod segments with undefected cladding.

The deionized water tests used fuel from the Turkey Point Unit 3 reactor and the J-13 groundwater tests were duplicated using both the Turkey Point and H. B. Robinson Unit 2 PWR fuels. Specimens were prepared from 5 inch long rod sections and the laser drilled, slit and undefected specimens were capped with water tight end fittings. Solution samples were periodically removed for analysis and replaced with fresh solution. Samples of fused quartz rod contained in the fused quartz test vessels were also periodically removed to monitor progression of radionuclide "plate-out". At the end of a test run (approximately 240 days), the test specimens were removed and rinsed and the test vessels were then stripped with 8 M HNO_3 . All samples, including rinse and strip solutions, were radiochemically analyzed.

Total measured release for several radionuclides were calculated by summation of release quantities indicated for each sample type. Total measured fractional release values were calculated by dividing the total measured release values by ORIGEN-2 calculated specimen inventories. Three notable observations based on total measured fractional release results⁽²⁾ from the first test runs in deionized and J-13 groundwater are:

- 1) The actinides appear to be released congruently in both water types.
- 2) Cs-137 and Tc-99 are released preferentially relative to the actinides in both water types.
- 3) Total measured fractional actinide release in the bare fuel tests was much greater than observed in the "slit defect" or "hole defects" test.

During the post-containment period there is a potential that the fuel structure could be altered by interaction with the air atmosphere expected to be present at that time. Thermodynamic data indicate that UO_2 should eventually oxidize through U_3O_8 stoichiometry to UO_3 in the post-containment repository environment. Oxidation to U_3O_8 would substantially degrade the fuel structure which may significantly increase its leachability by infiltrating groundwater. Lesser degrees of fuel oxidation may also increase leachability. Although thermodynamically favored, the rate of oxidation past the U_3O_7 stoichiometry appears to be too slow to directly measure on a practical laboratory time scale at post-containment relevant temperatures which are below about 130°C.

Spent fuel oxidation studies at 200°C and 225°C have shown that the oxidation rate up to U_3O_7 stoichiometry appears to be independent of particle size(3). Examination of partially oxidized spent fuel particles has indicated that the grain boundaries appear to have opened (possibly due to initial oxidation of fission products concentrated at the grain boundaries) providing paths for oxygen penetration of the fuel structure. The hypothesis of initial grain boundary oxidation is further supported by ion microprobe data on spent fuel partially oxidized in $^{18}O_2$. Additional testing is in progress which may allow prediction of spent oxidation behavior at lower temperatures relevant to the repository post-containment period.

REFERENCES

1. Code of Federal Regulations, "Disposal of High Level Nuclear Wastes in Geological Repositories - Licensing Procedures," Title 10, Chapter 1, Part 60, June 30, 1983 (Cited at 10 CFR 60).
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3. R. E. Einziger, R. E. Woodley, "Low Temperature Spent Fuel Oxidation Under Tuff Repository Conditions," paper presented at Waste Management 85 Conference, Tucson, Arizona, March 24-28, 1985, (HEDL-SA-3271).

